

THE COOL FLAMES EXPERIMENT

Howard Pearlman[†]
University of Southern California
Department of Aerospace and Mechanical Engineering
Los Angeles, CA 90089
howard.pearlman@grc.nasa.gov

Richard Chapek, Donna Neville, William Sheredy
NASA GRC, Cleveland, OH 44135

Ming-Shin Wu
NCMR, Cleveland, OH 44135

Robert Tornabene
ZIN Technologies, Brook Park, OH 44142

INTRODUCTION

A space-based experiment is currently under development to study diffusion-controlled, gas-phase, low temperature oxidation reactions, cool flames and auto-ignition in an unstirred, static reactor. At Earth's gravity (1g), natural convection due to self-heating during the course of slow reaction dominates diffusive transport and produces spatio-temporal variations in the thermal and thus species concentration profiles via the Arrhenius temperature dependence of the reaction rates. Natural convection is important in all terrestrial cool flame and auto-ignition studies, except for select low pressure, highly dilute (small temperature excess) studies in small vessels (i.e., small Rayleigh number).

On Earth, natural convection occurs when the Rayleigh number (Ra) exceeds a critical value of approximately 600 (Tyler, 1966; Fine, et. al., 1970; Barnard and Harwood, 1974). Typical values of the Ra, associated with cool flames and auto-ignitions, range from 10^4 - 10^5 (or larger), a regime where both natural convection and conduction heat transport are important. When natural convection occurs, it alters the temperature, hydrodynamic, and species concentration fields, thus generating a multi-dimensional field that is extremely difficult, if not impossible, to be modeled analytically (Griffiths, et. al., 1971; Melvin, 1969). This point has been emphasized recently by Kagan and co-workers (Kagan, et. al., 1997; Volpert, et. al., 2000) who have shown that explosion limits can shift depending on the characteristic length scale associated with the natural convection. Moreover, natural convection in unstirred reactors is never "sufficiently strong to generate a spatially uniform temperature distribution throughout the reacting gas (Griffiths et. al., 1974)." Thus, an unstirred, nonisothermal reaction on Earth does not reduce to that generated in a mechanically, well-stirred system.

Interestingly, however, thermal ignition theories (Semenov, 1958; Frank-Kamenetskii, 1939, 1940) and thermokinetic models (Yang, 1969) neglect natural convection and assume a heat transfer correlation of the form: $q = h(S/V)(\bar{T} - T_w)$ where q is the heat loss per unit volume, h is

[†] Corresponding Author

the heat transfer coefficient, S/V is the surface to volume ratio, and $(\bar{T} - T_w)$ is the spatially averaged temperature excess. This Newtonian form has been validated in spatially-uniform, well-stirred reactors, provided the effective heat transfer coefficient associated with the unsteady process is properly evaluated (Griffiths, et.al.,1974; Gray, et.al., 1974). Unfortunately, it is not a valid assumption for spatially-nonuniform temperature distributions induced by natural convection in unstirred reactors. "This is why the analysis of such a system is so difficult (Griffiths, et. al., 1974)."

Historically, the complexities associated with natural convection were perhaps recognized as early as 1938 (Frank-Kamenetskii, 1939) when thermal ignition theory was first developed. In the 1955 text *Diffusion and Heat Exchange in Chemical Kinetics*, Frank- Kamenetskii recognized that "the purely conductive theory can be applied at sufficiently low pressure and small dimensions of the vessel when the influence of natural convection can be disregarded." This was reiterated by Tyler in 1966 (Tyler, 1966) and further emphasized by Barnard and Harwood in 1974. Specifically, they state: "It is generally assumed that heat losses are purely conductive. While this may be valid for certain low pressure slow combustion regimes, it is unlikely to be true for the cool flame and ignition regimes (Barnard and Harwood, 1974)." While this statement is true for terrestrial experiments, the purely conductive heat transport assumption is *valid* at microgravity (μg).

Specifically, buoyant complexities are suppressed at μg and the reaction-diffusion structure associated with low temperature oxidation reactions, cool flames and auto-ignitions can be studied (Pearlman, 1999, 2000). Without natural convection, the system is simpler, does not require determination of the effective heat transfer coefficient, and is a testbed for analytic and numerical models that assume pure diffusive transport. In addition, μg experiments will provide baseline data that will improve our understanding of the effects of natural convection on Earth.

GROUND-BASED COOL FLAME AND AUTO-IGNITION STUDIES

Laboratory (1g) and KC-135 aircraft experiments are actively being conducted in preparation for the space flight. These tests are performed in a spherical, quartz reactor, housed in a preheated furnace. The vessel is filled with a fuel-oxidizer premixture and the ensuing reaction is monitored. Details of the hardware are discussed in the references 10 and 11.

Cool Flames

Representative cool flame images obtained in the lab and aboard the KC-135 aircraft are shown in figures 1a and 1b. Note that the cool flame at μg starts in the center of the spherical reactor and propagates radially outward.

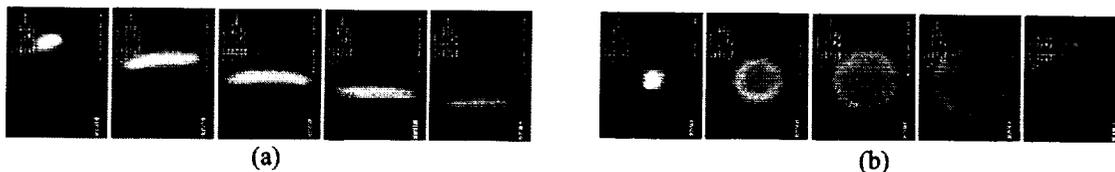


Fig.1: Sequential cool flames images obtained using a 50v%n-C₄H₁₀-50v%O₂ premixture at 310°C and 3.2psia in a 10cm i.d. spherical quartz reactor in the (a) 1g and (b) KC-135 μg aircraft.

transport times at μg (diffusion times $\sim L^2/\alpha$ and L^2/D , where α is the mixture thermal diffusivity and D is species diffusion coefficient) compared to 1g (buoyant rise time $\sim (gL)^{1/2}$).

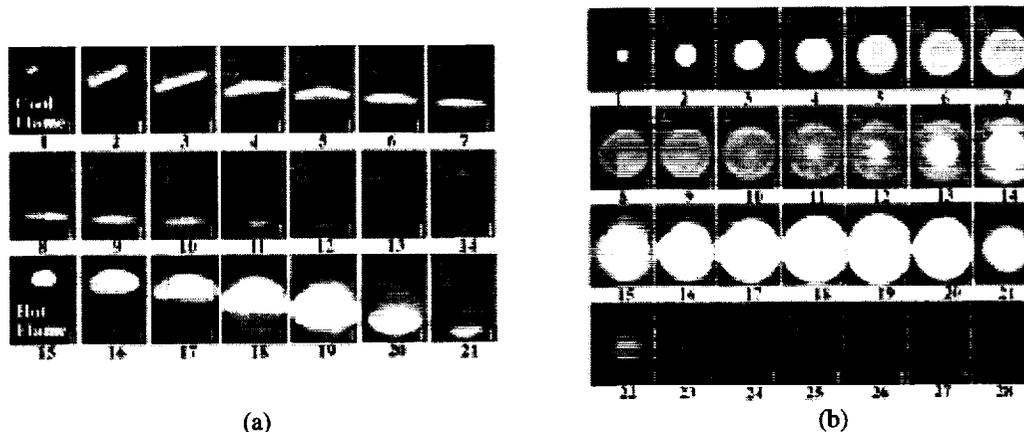


Fig. 4: Two-Stage Ignition in 50v% n-C₄H₁₀-50v% O₂ mixture, $P_{\text{initial}}=4.9\text{psia}$, $T=310^\circ\text{C}$, Vessel i.d.=10.2cm, (a) 1g, Time between sequential frames is 1/10s, (b) μg ; Time between sequential frames is 1/30s.

Single-stage ignition at μg has also been observed. Future effort aims to quantify the flame structure, evolution, and stability and map the ignition diagrams for different hydrocarbons.

ACKNOWLEDGMENTS

Special thanks to Professor John Griffiths for helpful discussions. This work is supported by NASA under NCC3-871.

REFERENCES

1. Barnard, J.A. and Harwood, B.A. (1974) *Combustion and Flame* **22** 35-42.
2. Fine, D.H., Gray, P. and MacKinven, R. (1970) *Royal Society of London* **A316** 223-240.
3. Frank-Kamenetskii, D.A. (1939) *Zh. Fiz. Khim.* **13** 738.
4. Frank-Kamenetskii, D.A. *Diffusion and Heat Exchange in Chemical Kinetics*, Princeton University Press, 1955.
5. Gray, P., Griffiths, J.F., and Moule, R.J. (1974) *Faraday Symposia of the Chemical Society* **9** 103-113.
6. Griffiths, J.F., Gray, B.F., and P. Gray (1971) *13th Symposium (Int.) on Comb.*, 239-248.
7. Griffiths, J.F., Gray, P., and Kishore, K. (1974) *Combustion and Flame* **22** 197-207.
8. Kagan, L., Beresycki, H., Joulin, G. and Sivashinsky, G. (1997) *CTM* **1** 97-111.
9. Melvin, A. (1969) *Combustion and Flame* **13** 438-9.
10. Pearlman, H. (1999) *Combustion and Flame* **121** (1-2) 390-3.
11. Pearlman, H. *The 3rd International Seminar on Fire and Explosion Hazards of Substances*, Lake Windermere, UK, April 10-14, 2000.
12. Semenov, N. N. *Some Problems of Chemical Kinetics and Reactivity*, Vol. 1, Pergamon Press, New York, 1958.
13. Tyler, B.J. (1966) *Combustion and Flame* **10** 90-91.
14. Volpert, V., Barillon, C., Dumont, T., Genieys, S., and Massot, M, *The Fire and Explosion Conf. hosted by the Univ. of Central Lancashire*, Windermere, UK, August 10-14, 2000.
15. Yang, C.H. and Gray, B.F. (1969) *Journal of Physical Chemistry* **73** (10) 3395-3406.

Multiple Cool Flames

Multiple cool flames are also observed at 1g for a select range of test parameters. To date, however, only one cool flame has been observed in the butane-oxygen system at μg , within the available 23s test time. To determine if the absence of multiple cool flames at μg is due to insufficient test time for thermal relaxation, an equimolar mixture of $n\text{-C}_4\text{H}_{10}\text{-O}_2$ was diluted with equal volumes of helium or argon to vary the thermal diffusivity of the mixture ($\alpha_{\text{He-mix}} \sim 4\alpha_{\text{Ar-mix}}$), thus, adjust the thermal relaxation time ($t_{\text{th}} \sim L^2/\alpha$).

The pressure histories associated with the reaction in the (a) Ar and the (b) He - diluted mixtures at μg are shown in figures 2a and 2b, respectively. The first pressure peak in the Ar-diluted trace and all pressure peaks in the He-diluted trace correspond to cool flames, as verified with the video record. As many as five sequential cool flames have been observed at μg !

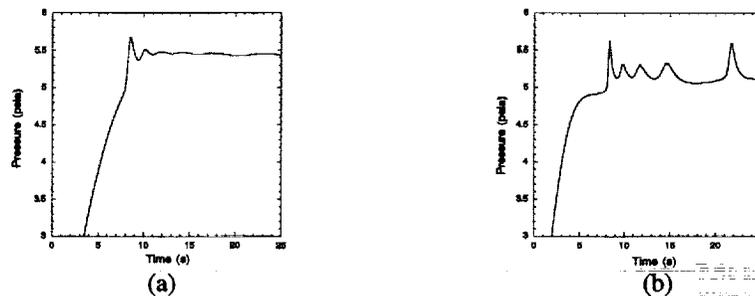


Fig. 2: Pressure traces at μg for a 25% $n\text{-C}_4\text{H}_{10}\text{-25}\%\text{O}_2$ mixture diluted with (a) 50%Ar, (b) 50%He in an aged 10.2cm i.d. spherical vessel, $T_{\text{vessel}}=310^\circ\text{C}$, $P_{\text{initial}}=4.95\text{psia}$.

Interestingly, the magnitude of the pressure excursions associated with successive cool flames in the He-diluted mixture at μg do not vary monotonically. As one might also expect, the intensity (integrated spectral emission) also varies. This is shown in figure 3, which corresponds to four successive cool flames in a He-diluted mixture at μg .

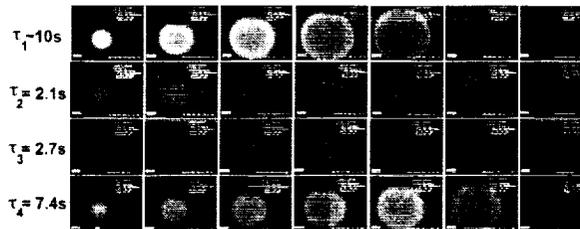


Fig.3: Multiple cool flames at μg ; τ_i ($i=1,2,3,4$) are the induction times; premixture: 25% $n\text{-C}_4\text{H}_{10}\text{-25}\%\text{O}_2\text{-50}\%\text{He}$ in an aged 10.2cm i.d. spherical vessel, $T_{\text{vessel}}=310^\circ\text{C}$, $P_{\text{initial}}=4.2\text{psia}$.

Multi-Stage Ignition

Multi-stage ignition, one (or more) cool flames followed by a hot ignition, has also observed at 1g and μg . Representative side-view images, depicting the development and progression of a representative two-stage ignition are shown in figures 4a and 4b, at 1g and μg , respectively. Similar to the cool flames shown in figure 1, both the cool flame and hot flame associated with the two-stage process originate near the top of the vessel at 1g and near the center at μg . Note that the second induction period (the time between the cool flame and the hot flame) is also shorter at μg than at 1g, perhaps due to the shorter